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EXCHANGE COUPLING AND SPIN-FLIP TRANSITION OF $CoFe_2O_4/\alpha$-Fe_2O_3 BILAYERED FILMS

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ABSTRACT

$CoFe_2O_4/\alpha$-Fe_2O_3 (ferrimagnetic / antiferromagnetic) bilayered films were prepared on α-Al_2O_3(102) single-crystalline substrates by helicon plasma sputtering. A well-crystallized epitaxial α-Fe_2O_3(102) layer was formed on the substrate, while $CoFe_2O_4$ grown on α-Fe_2O_3 (102) was a polycrystalline layer with a (100)-preferred orientation. The α-Fe_2O_3(102) films without $CoFe_2O_4$ layers clearly showed a spin-flip transition at about 400 K. The spins aligned perpendicular to the film plane at room temperature changed their direction within the film plane above 400 K. However the α-Fe_2O_3 base layers of $CoFe_2O_4/\alpha$-Fe_2O_3 bilayered films did not show any spin-flip transition. The $CoFe_2O_4$ layer on α-Fe_2O_3 had a large in-plane magnetic anisotropy, while the spin axis of the α-Fe_2O_3 (102) base layer was directed perpendicular to the film plane. The magnetization of ferrimagnetic $CoFe_2O_4$ layers was coupled perpendicularly to the spin axis of anitiferromagnetic α-Fe_2O_3 layers due to the exchange coupling at the interface between $CoFe_2O_4$ and α-Fe_2O_3.

INTRODUCTION

Exchange coupling at an interface between ferromagnetic (FM) and antiferromagnetic (AFM) layers has received much attention mainly due to the technological applications in such devices as spin-valve sensors. It stabilizes a magnetic direction of the FM layer and functions as a bias field in the magnetic hysteresis loop. Recently, full micromagnetic calculations suggested the existence of the 90° FM - AFM coupling at the interface [1]. However the fundamental origin of exchange coupling between magnetic materials, especially magnetic oxide materials, is still unclear. α-Fe_2O_3 is one of candidates for the AFM materials fabricated in the spin-valve sensors [2,3]. The spin valves partly consisting of α-Fe_2O_3 have high thermal stability and large magneto-resistance ratio. By the way, one of the present authors found that epitaxial α-Fe_2O_3 (102) films on α-Al_2O_3(102) had an unique spin-flip transition [4]. The transition takes place at about 400 K, much higher than the Morin transition temperature (260 K) of the bulk crystal. The spin axis lying within a film plane above 400 K turns perpendicular to the film plane below the transition temperature.

There are a few studies on the exchange coupling between oxide materials with the FM/AFM bilayered structure [5]. Most of practical FM oxides are exactly ferrimagnetic. Microscopic spin configurations at the interface between oxide systems could be different from the ones between metallic systems. We prepared well-crystallized α-Fe_2O_3 and $CoFe_2O_4$ bilayered films by using helicon plasma sputtering technique [6]. $CoFe_2O_4$ is a typical ferrimagnetic material with an inverse spinel structure. Structural properties and magnetic interactions between ferrimagnetic $CoFe_2O_4$ (FM) and α-Fe_2O_3 (AFM) layers were discussed. If the exchange coupling at the $CoFe_2O_4/\alpha$-Fe_2O_3 interface was strong enough, magnetic properties of the $CoFe_2O_4$ layer should be influenced by the spin-flip transition of the α-Fe_2O_3 layer or vice versa.

EXPERIMENT

Helicon plasma sputtering is a powerful technique to prepare high-quality multilayered films with sharp interfaces [7]. It has some advantages in comparison with conventional rf magnetron sputtering, such as high deposition rate stability and low plasma damage to the film surface. A helicon plasma cathode consists of a conventional rf magnetron cathode and a rf coil for a helicon wave. Fig.1 shows a schematic drawing of the helicon plasma sputtering system we used. Two targets for helicon cathodes were made of sintered α-Fe_2O_3 and CoO, respectively. The base pressure of the system was 10^{-7} Pa. Before sputter deposition an α-Al_2O_3(102) substrate was annealed in vacuum at about 973 K for 1 hour in order to obtain a clean and well-ordered surface. α-Fe_2O_3 base layers with the thickness of 100 nm were sputtered on the substrate at the substrate temperature of 673 K. $CoFe_2O_4$ layers with several thicknesses ranging from 25 to 200 nm were then deposited on α-Fe_2O_3 at 773 K by simultaneous sputtering from both targets, to control the deposition rate ratio between α-Fe_2O_3 and CoO. The deposited films were characterized by reflection high energy electron diffraction (RHEED), x-ray diffraction (XRD), scanning probe microscopy (SPM), vibrating sample magnetometer (VSM), conversion electron Mössbauer spectroscopy (CEMS), and energy dispersive X-ray spectroscopy (EDS).

RESULTS AND DISCUSSION

Typical XRD and RHEED patterns of α-Fe_2O_3 films deposited on α-Al_2O_3 (102) single-crystalline substrates are shown in figs. 2(a) and (b), respectively. Epitaxial relationship between the α-Fe_2O_3 layer and the α-Al_2O_3 substrate can clearly be seen in both XRD and RHHED patterns. The XRD pattern had only a reflection from the film indexed as α-Fe_2O_3(204) at the side of an intense α-Al_2O_3 (204) reflection. The sharp streak lines in the RHEED pattern indicated an atomically flat surface of the layer. Both α-Fe_2O_3 and α-Al_2O_3 have a corundum structure with a small lattice misfit of +5.8 %. This could be a reason why the well-crystallized and atomically flat α-Fe_2O_3 layers were epitaxially formed on the α-Al_2O_3 substrates.

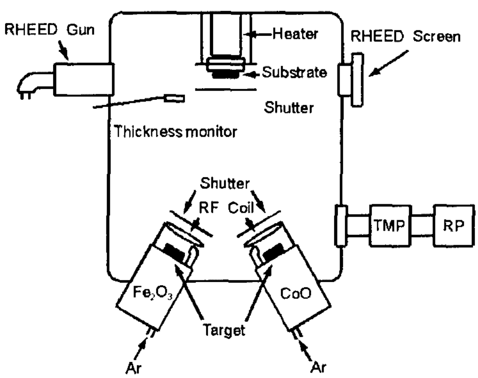

Figure 1. Schematic drawing of helicon plasma sputtering system we used. The helicon cathodes consist of conventional rf magnetron cathodes and rf coils.

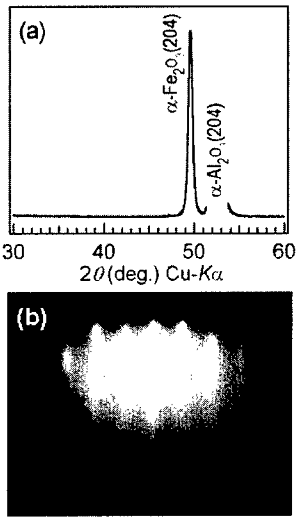

Figure 2. (a) XRD and (b) RHEED patterns of an α-Fe_2O_3 film deposited on an α-Al_2O_3 (102) substrate.

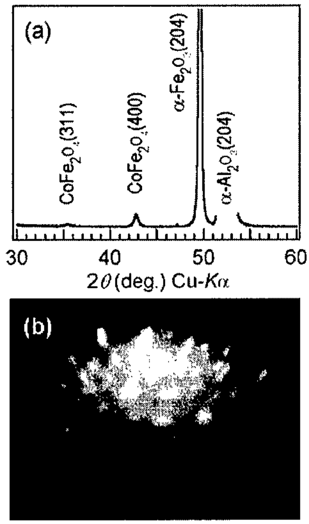

Figure 3. (a) XRD and (b) RHEED patterns of a $CoFe_2O_4$/α-Fe_2O_3 bilayered film deposited on an α-Al_2O_3 (102) substrate. Thicknesses of the $CoFe_2O_4$ and the α-Fe_2O_3 layers were 200 and 100 nm, respectively.

Figs. 3(a) and (b) are typical XRD and RHEED patterns of $CoFe_2O_4/Fe_2O_3$ bilayered films, respectively. The XRD pattern of the $CoFe_2O_4/Fe_2O_3$ bilayered film had two reflections from the $CoFe_2O_4$ layer indexed as $CoFe_2O_4$(311) and (400) in addition to the reflection from the α-Fe_2O_3 (204) base layer. The relative peak intensity ratio of $CoFe_2O_4$(400) to (311) was considerably large, in comparison with that of the $CoFe_2O_4$ bulk pattern with random orientation [8]. The $CoFe_2O_4$ layer formed on α-Fe_2O_3(102) had strong (100)-preferred orientation. In crystallographic aspects, the α-Fe_2O_3(102) surface has a pseudo-square structure though the α-Fe_2O_3 crystal has hexagonal corundum structure. The $CoFe_2O_4$(100) layer with cubic spinel structure could be formed on the α-Fe_2O_3 (102) base layer. However the lattice misfit between them is very large, about −17%. The RHEED pattern of $CoFe_2O_4$ on α-Fe_2O_3(102) was spotted and complicated. The $CoFe_2O_4$ layers in bilayered films were poly-crystallized and had (100)-preferred orientation. Moreover, chemical formula of the $CoFe_2O_4$ layer analyzed by EDS was $Co_{0.7}Fe_{2.3}O_4$.

The spin direction of α-Fe_2O_3 layers on α-Al_2O_3(102) was easily determined by CEMS. The CEMS spectrum of α-Fe_2O_3 generally exhibited six lines due to the nuclear Zeeman splitting from a large internal magnetic field. Relative peak intensity ratio of the sextet is expressed theoretically as a function of an angle (θ) between the γ-ray direction and the spin direction

$$3:\frac{4\sin^2\theta}{1+\cos^2\theta}:1:1:\frac{4\sin^2\theta}{1+\cos^2\theta}:3.$$

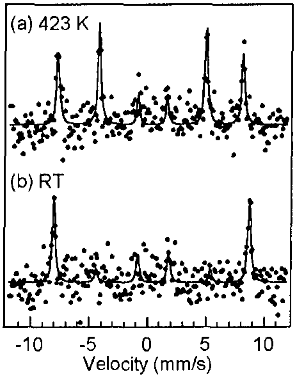

Figure 4. *CEMS spectra of an α-Fe_2O_3(102) film deposited on α-Al_2O_3 (102) measured at (a) 423 K and (b) room temperature.*

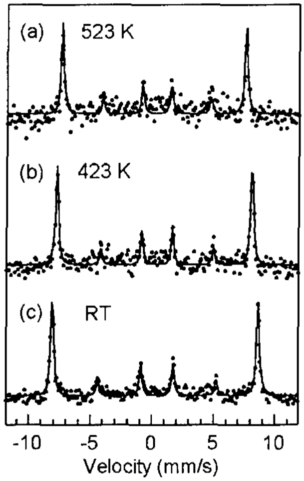

Figure 5. *CEMS spectra of a $CoFe_2O_4$/α-Fe_2O_3 bilayered film on α-Al_2O_3 (102), measured at (a) 523 K, (b) 423 K, and (c) room temperature. Thicknesses of the $CoFe_2O_4$ and α-Fe_2O_3 layers were 25 and 100 nm, respectively.*

CEMS spectra of an α-Fe_2O_3(102) film without a $CoFe_2O_4$ covering layer are shown in fig. 4 as a function of the temperature. The spectra had the intensity ratio of nearly 3:0:1:1:0:3 at 300 K and 3:4:1:1:4:3 at 423 K. The spin axis of the α-Fe_2O_3 (102) film was abruptly changed from the perpendicular direction (θ=0°) to the in-plane direction (θ=90°) at about 400 K.

Besides the intensity ratio of the CEMS spectra of the $CoFe_2O_4$/α-Fe_2O_3(102) bilayered film did not show any temperature dependence, even when the α-Fe_2O_3(102) layer was covered by a very thin, 25 nm-thick, $CoFe_2O_4$ layer. All spectra of the α-Fe_2O_3(102) base layer had the intensity ratio of about 3:0:1:1:0:3 as shown in fig. 5. The α-Fe_2O_3 layer covered by $CoFe_2O_4$ did not show the spin-flip transition. The spin direction in α-Fe_2O_3 was fixed perpendicular to the film plane over the all temperatures.

In-plane and out-of-plane magnetization curves of the $CoFe_2O_4$/α-Fe_2O_3 bilayered films were also measured at various temperatures. Figs. 6(a) and (b) show room temperature magnetization curves of the 200 nm-thick $CoFe_2O_4$ films without and with the α-Fe_2O_3(102) base layers, respectively. No uniaxial magnetic anisotropy nor exchange bias field was induced in the $CoFe_2O_4$ layers, when they did not have the α-Fe_2O_3 base layers. $CoFe_2O_4$ is known to have a large magnetocrystalline anisotropy along the <100> direction [8]. The (100)-oriented $CoFe_2O_4$ layers could have domain structures magnetized along the in-plane [100] and [010] and the out-of-plane [001] directions. The large coercivity was, thus, observed in both in-plane and out-of-plane hysteresis loops.

On the other hands, the $CoFe_2O_4$ layers deposited on the α-Fe_2O_3(102) base layers had a large in-plane magnetic anisotropy. The exchange bias field of about 200 Oe was induced. The spin

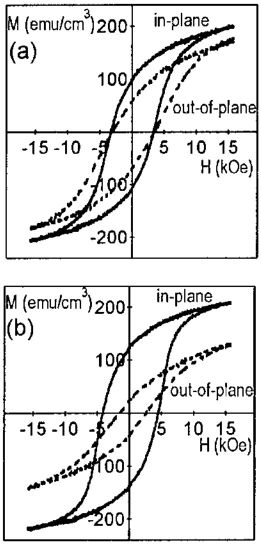

Figure 6. *In-plane and out-of-plane magnetization curves of (a) a 200 nm-thick $CoFe_2O_4$ film deposited on α-Al_2O_3(102), and (b) a $CoFe_2O_4$ (200 nm) /α-Fe_2O_3 (100 nm) bilayered film on α-Al_2O_3 (102). The magnetization curves were measured at room temperature*

axis of the α-Fe_2O_3 base layers was directed perpendicular to the film plane as discussed above. The magnetization of the $CoFe_2O_4$ (FM) layers was exactly coupled perpendicularly to the spin axis of the α-Fe_2O_3 (AFM) layers. The 90° FM-AFM coupling observed in $CoFe_2O_4$/α-Fe_2O_3 bilayered films was in good agreement with the theoretical result reported by Koon [1]. The large in-plane anisotropy of the $CoFe_2O_4$ layers was probably induced by the 90° coupling with the α-Fe_2O_3 layers and it should suppress the spin-flip transition of the α-Fe_2O_3 layers irreversibly.

SUMMARY

Exchange coupling between a ferrimagnetic $CoFe_2O_4$ layer and an anitiferromagnetic α-Fe_2O_3 layer was examined to prepare $CoFe_2O_4$/α-Fe_2O_3 bilayered films. (100)-oriented $CoFe_2O_4$ layers were formed on well-crystallized epitaxial α-Fe_2O_3(102) layers deposited on α-Al_2O_3(102) single-crystalline substrates. The α-Fe_2O_3(102) films without $CoFe_2O_4$ layers clearly showed the spin-flip transition at about 400 K. The spin axis lay within the (102) plane of α-Fe_2O_3 at high temperatures but became almost normal to the plane at low temperatures. However the α-Fe_2O_3(102) layers covered by $CoFe_2O_4$ layers did not show any spin-flip transition. The spin axis of the α-Fe_2O_3(102) base layers was fixed on the perpendicular to the films. Magnetic hysteresis loops of $CoFe_2O_4$/α-Fe_2O_3 bilayered films indicated that a large in-plane magnetic anisotropy and an exchange bias field were induced in the films. The 90° coupling at the interface between ferrimagnetic and antiferromagnetic materials was directly observed in the $CoFe_2O_4$/α-Fe_2O_3 system fabricated entirely of oxides.

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